Probing two-alpha radioactivity with ^{224}Ra

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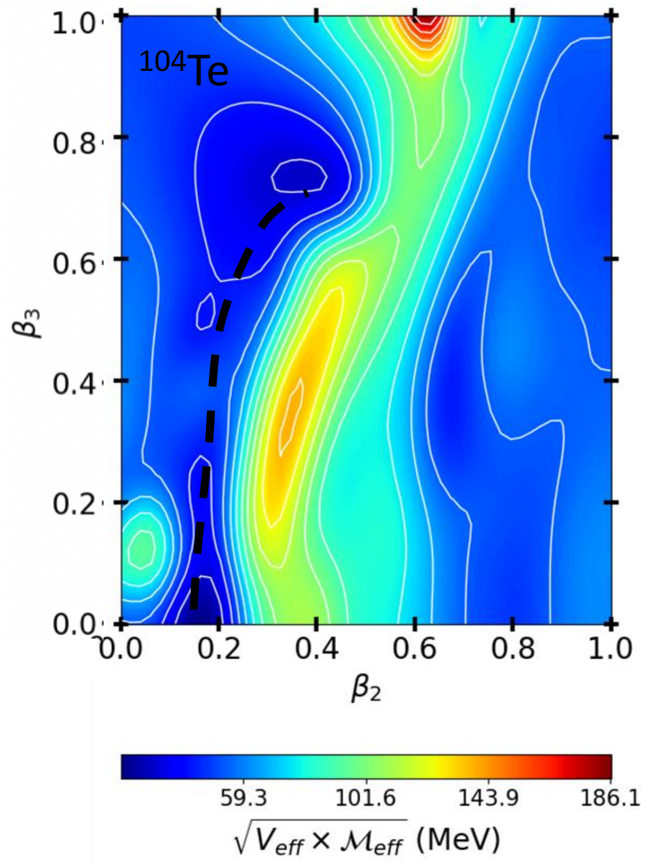
A microscopic approach to α decay

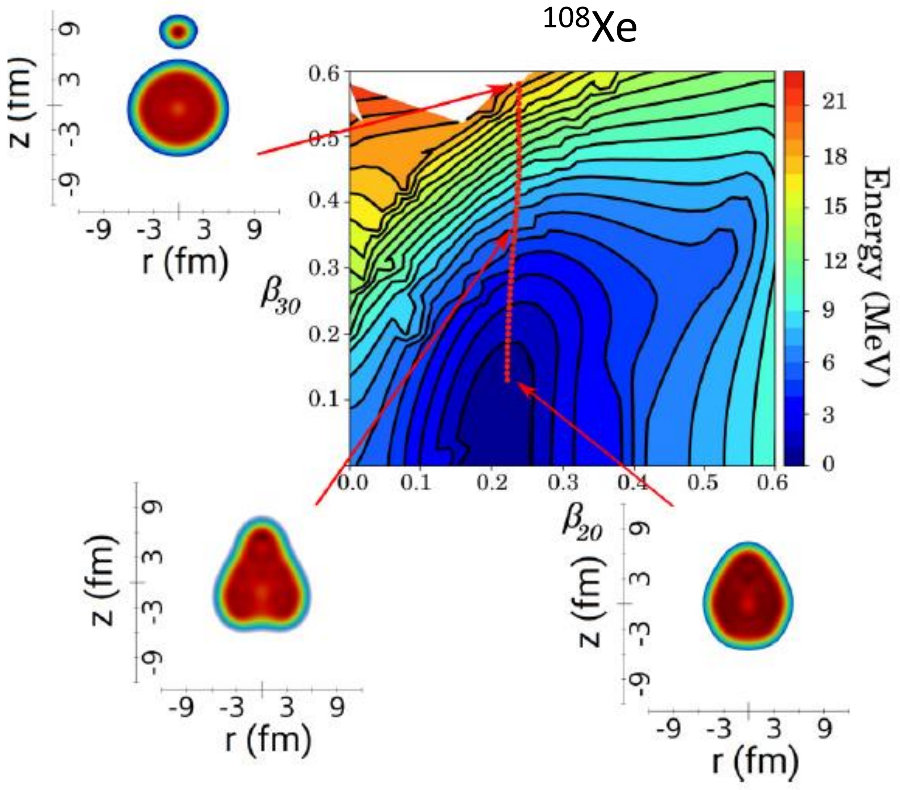
Potential energy surfaces calculated with covariant EDF

$$S(L) = \int_{s_{in}}^{s_{out}} \frac{1}{\hbar} \sqrt{2\mathcal{M}_{eff}(s)[V_{eff}(s) - E_0]}\, ds$$: minimization of the action integral

$$P = \frac{1}{1 + \exp[2S(L)]}$$: barrier penetration probability (WKB)

$$T_{1/2} = \ln 2/(nP)$$ n: number of assaults per unit of time

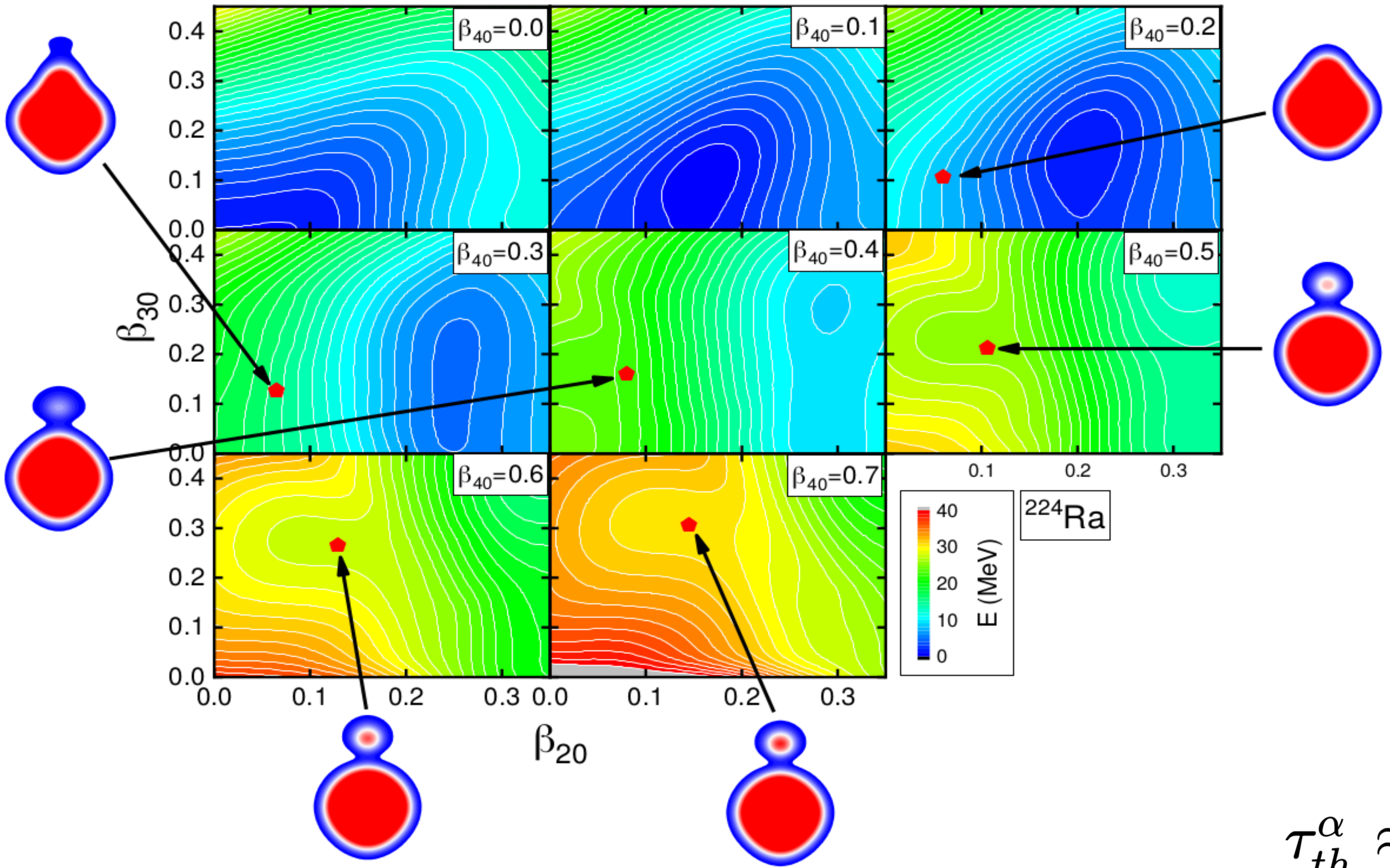


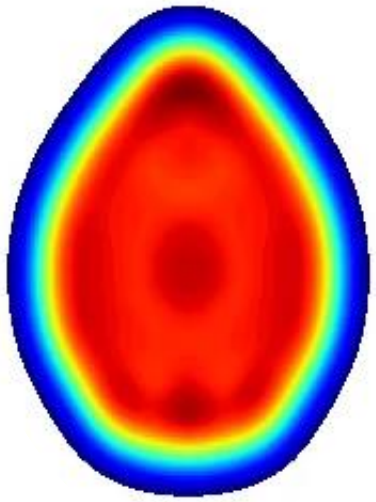


	^{104}Te	^{108}Xe	^{212}Po	^{224}Ra
T_{exp}	<18ns	58 μs	0.3 μs	3.6 d
T_{theo}	197ns	50 μs	0.6 μs	9.5 d

F. Mercier et al., Phys. Rev. C 102, 011301(R) (2020)
Phys. Rev. Lett. 127, 012501 (2021)

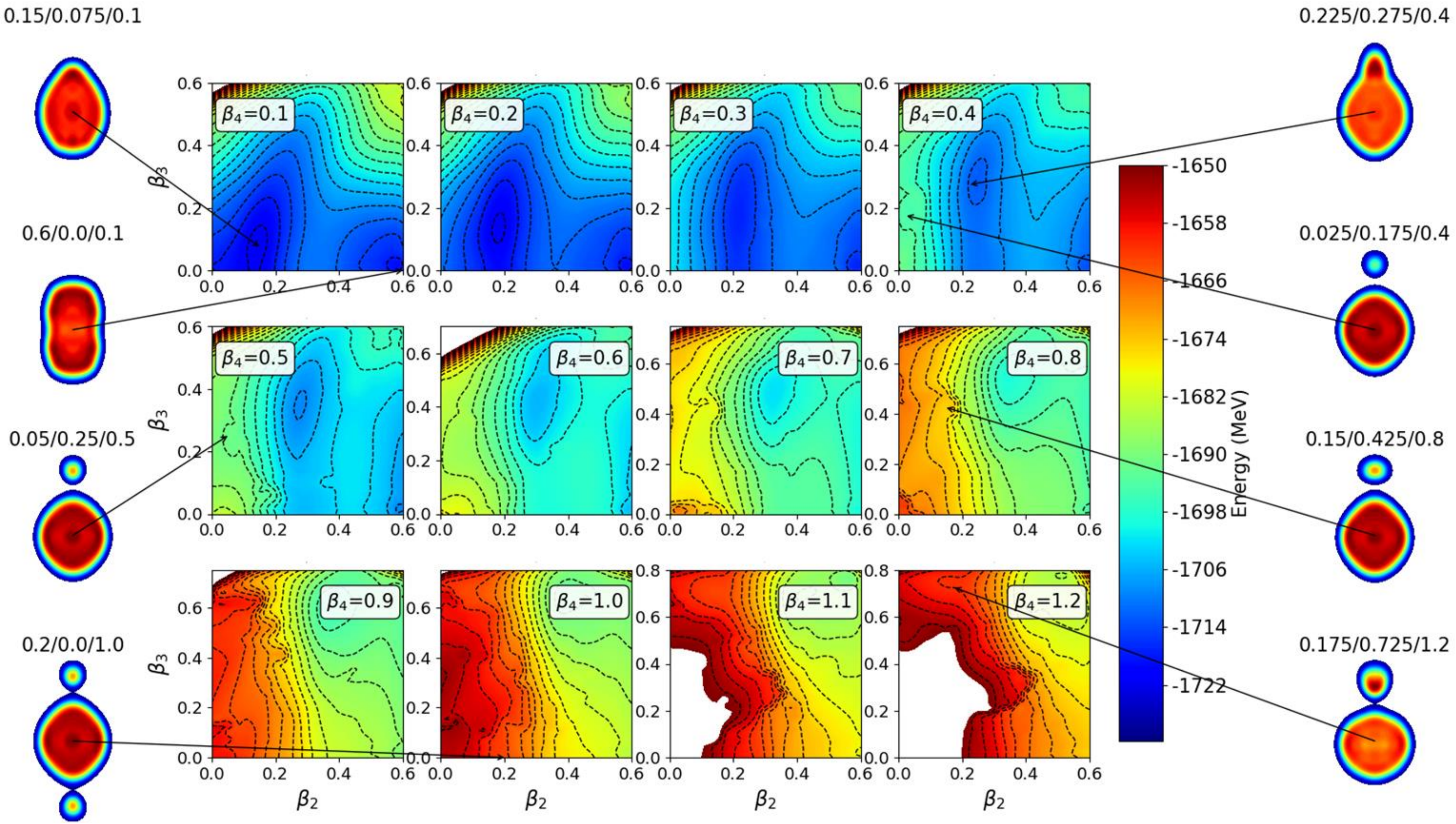
α decay in ^{224}Ra



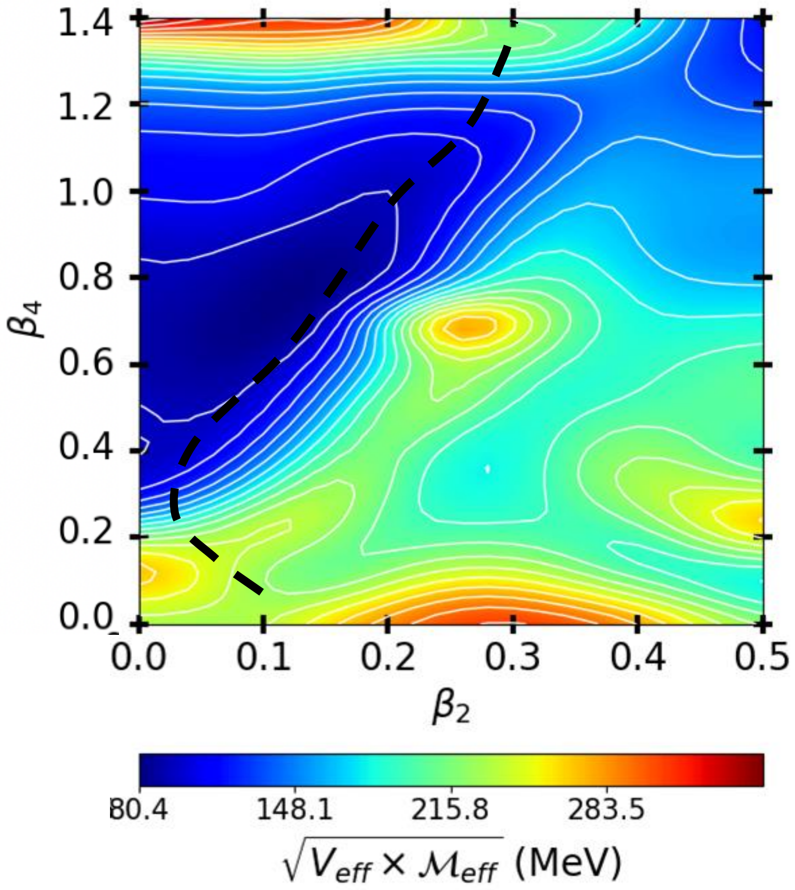


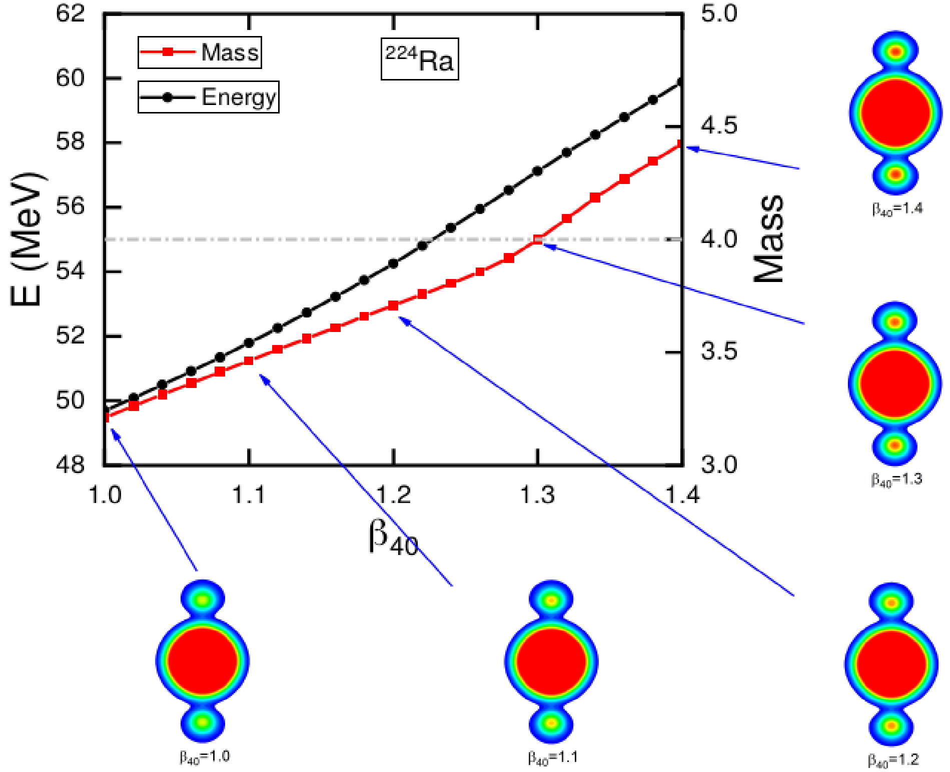
$$\tau^{\alpha}_{th} \approx 8.2 \times 10^5 \text{ s}$$
$$\tau^{\alpha}_{exp} \approx 3.1 \times 10^5 \text{ s}$$

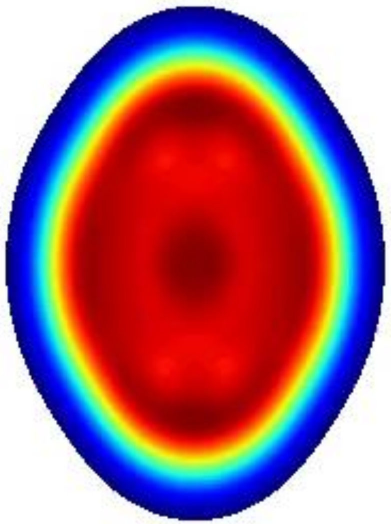
3D PES in ^{224}Ra



2α decay in ^{224}Ra







	^{14}C	^{8}Be	2α
T_{theo}	$10^{17.46}$ s	$10^{27.87}$ s	$10^{14.24}$ s
T_{exp}	$10^{15.86}$ s	??	??

Experimental considerations

- Predicted Branching Ratio = $T_{1/2}(\alpha)/T_{1/2}(2\alpha) \sim 10^{-9}$

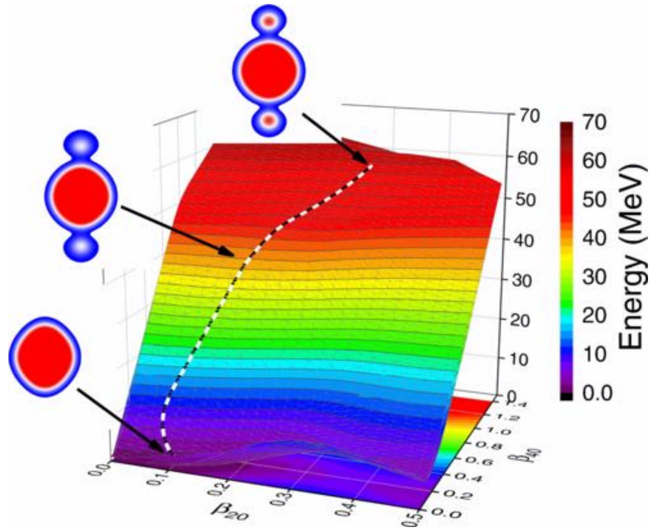
→ at least 10^{10} ^{224}Ra needed

→ Back to back detection of two alphas

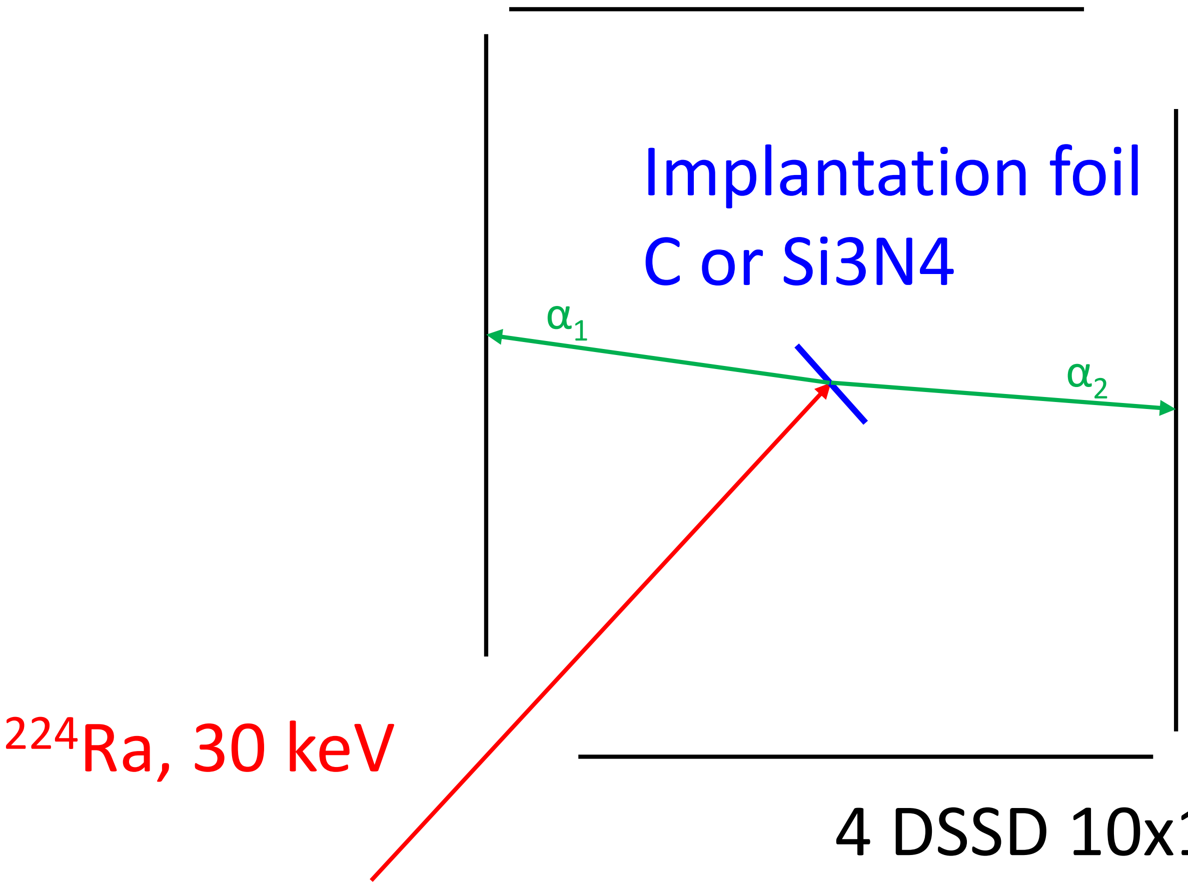
- Alpha energies: one expects the energy to be almost equally shared between the two alphas (reflection symmetric path)

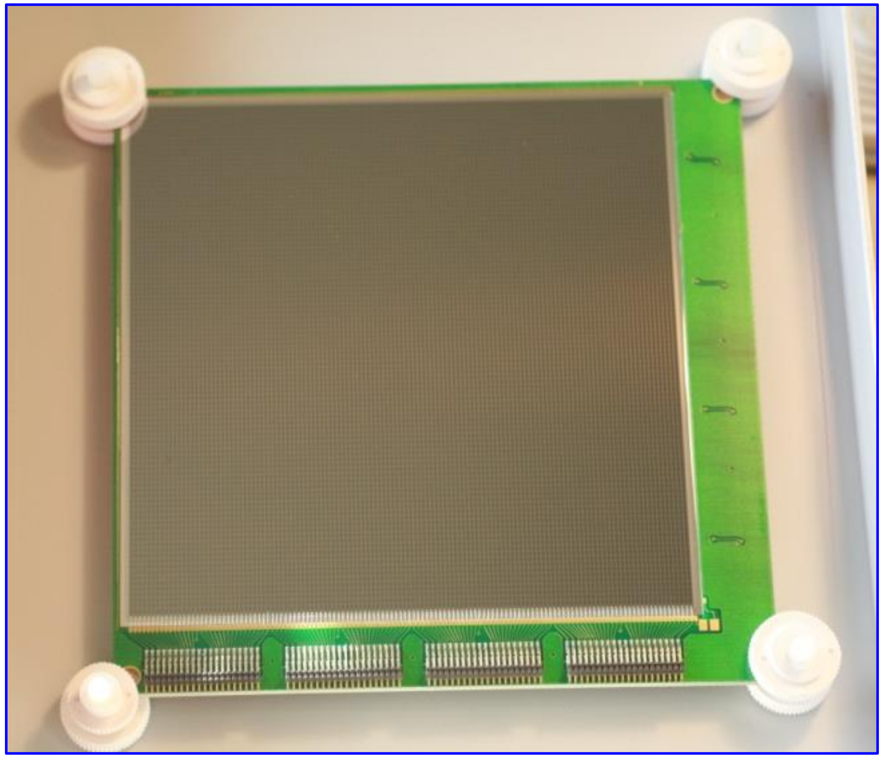
→ $E_{sum} = E_{2}\alpha(^{224}Ra) \sim Q\alpha(^{224}Ra) + Q\alpha\ (^{220}Rn) \sim 12.19$ MeV

Kinematic correction will be done when $E_{\alpha 1} \neq E_{\alpha 2}$



Detection principle





4 DSSD 10x10 cm^2

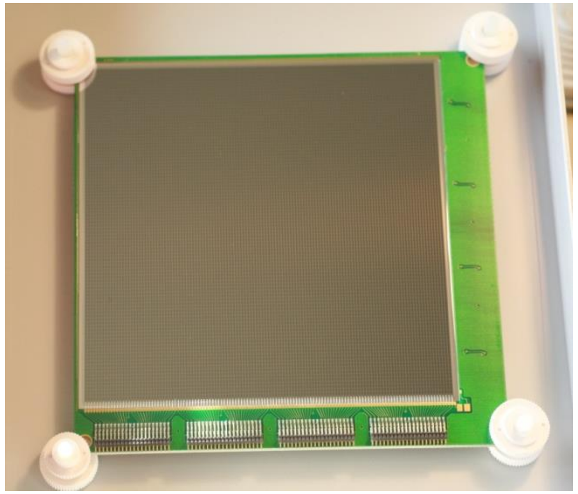
Foil – DSSD ~ 6 cm

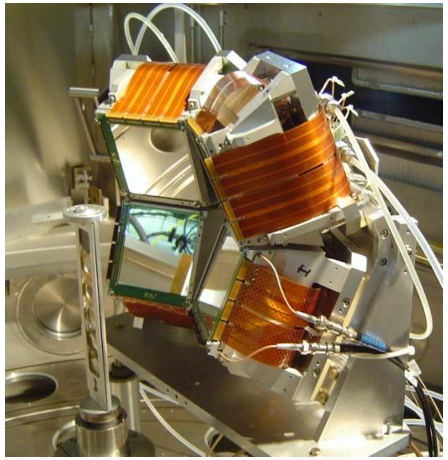
Detection efficiency ~ 45%

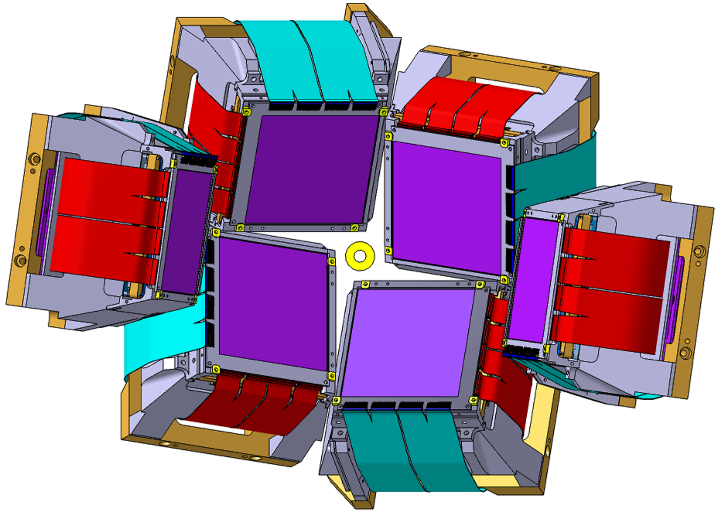
C 200 nm / Si3N4 100 nm

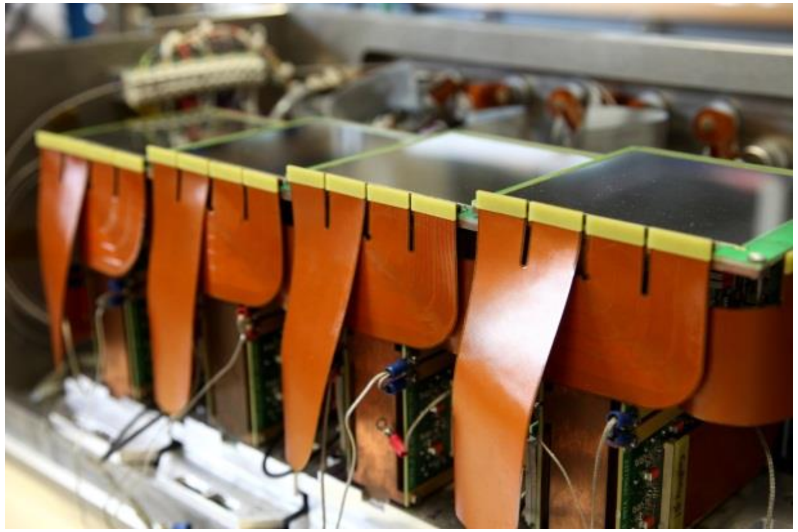
The MUST II and MUSETT arrays

- 10x10 cm^2 DSSDs 128+128 strips, 300µm
- Versatile geometry





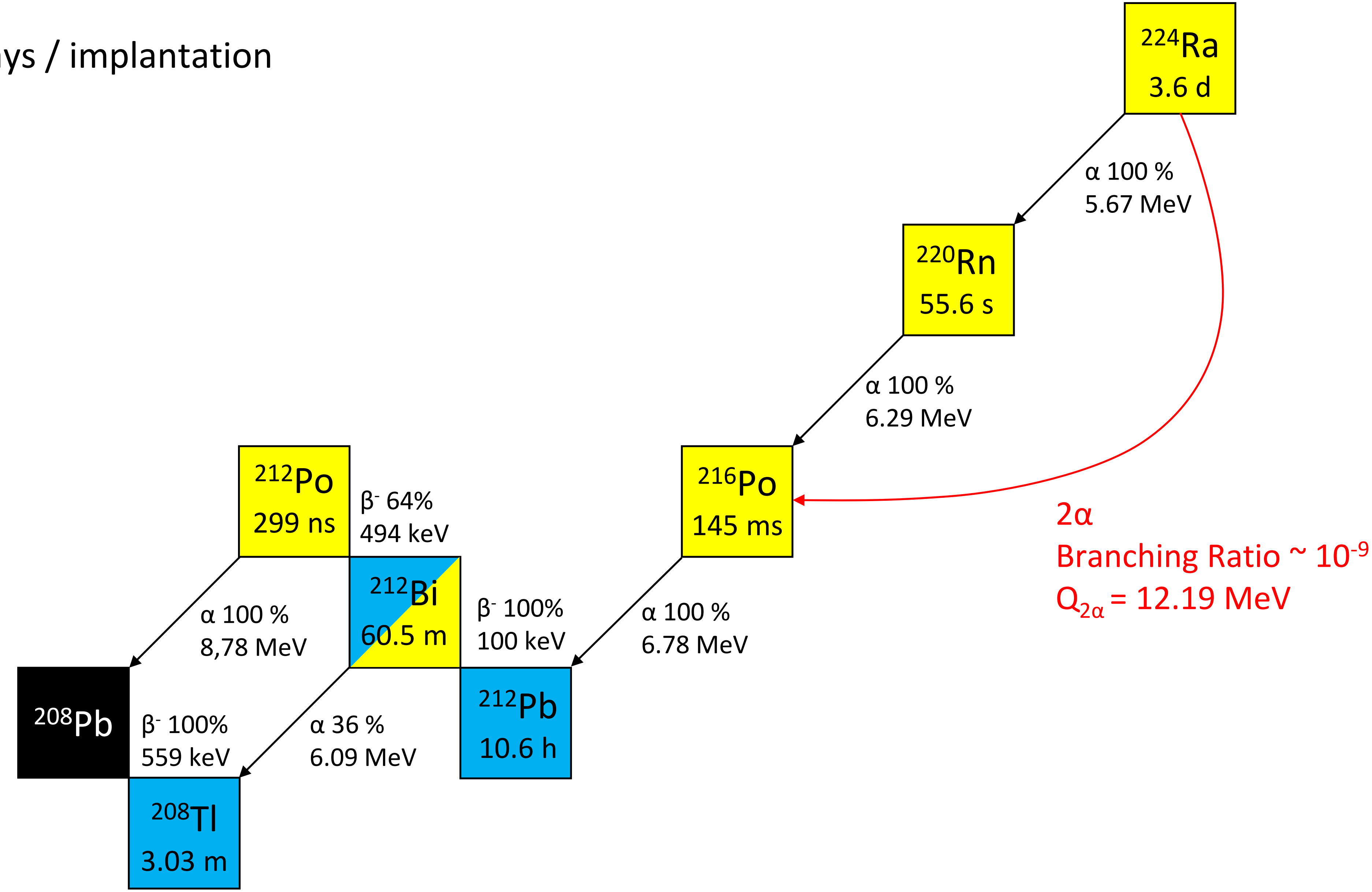




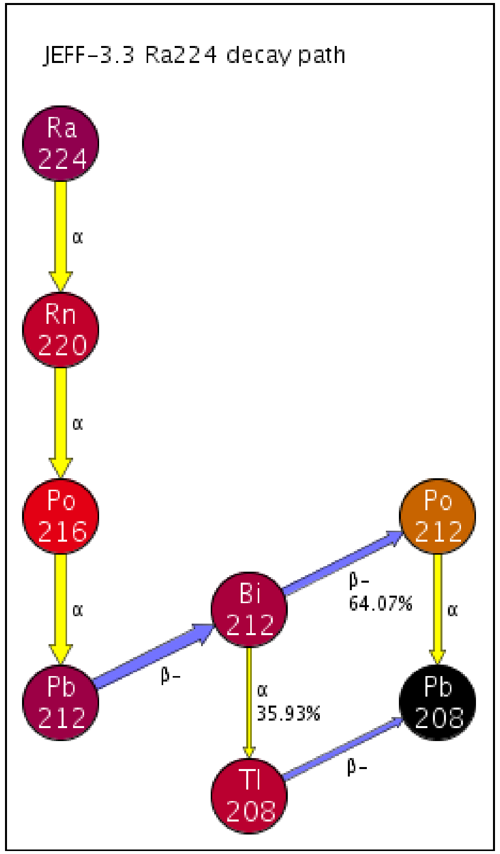
- New MUSETT/MUST II geometry
- Setup in LA1 line
- Reaction chamber + pumps
- 1 electronics rack (VXI, NIM, Caen HT+BT)
- Cryo-cooler
- GANIL standalone DAQ

^{224}Ra decay chain

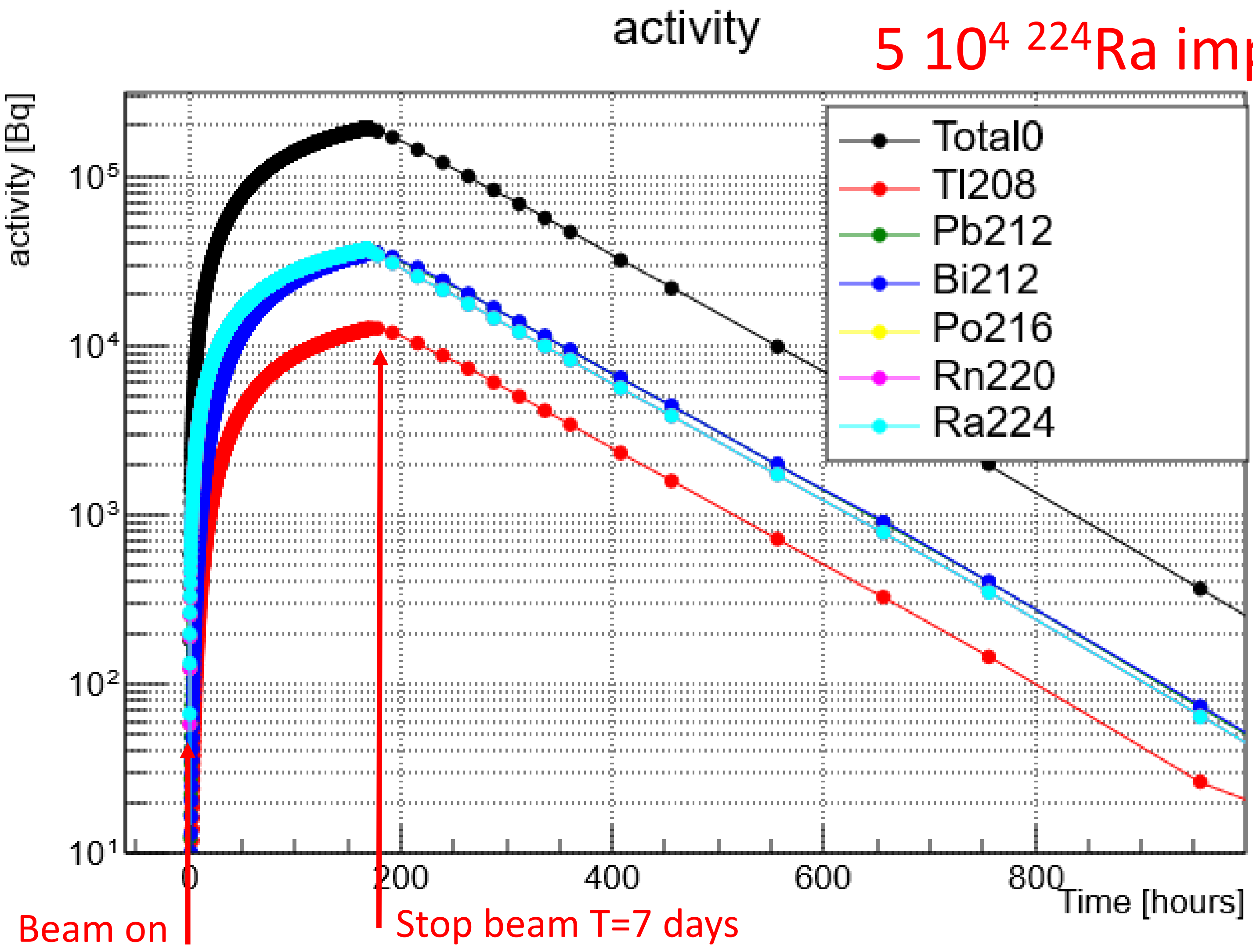
- 6 decays / implantation



beam time + offline



$5\ 10^4\ {}^{224}Ra$ implantations/s



~ 13 double alpha expected (emitted)

~ 9 (13) double alpha in 4 (7) days (emitted)

FISPACT-II code (NEA)
Courtesy L. Thulliez Irfu/DPhN

Random coincidences, selection, etc.

$5\ 10^4$ ^{224}Ra implantations/s → $3\ 10^{10}$ in one week.

- Assume activity = $3\ 10^5$ Bq (most pessimistic case)
- β-decay can by partly suppressed by electronics threshold

→ Total detection rate ~ 10^5 Hz (most pessimistic case)

→ Random coincidence rate in a 100 ns window = 350 Hz

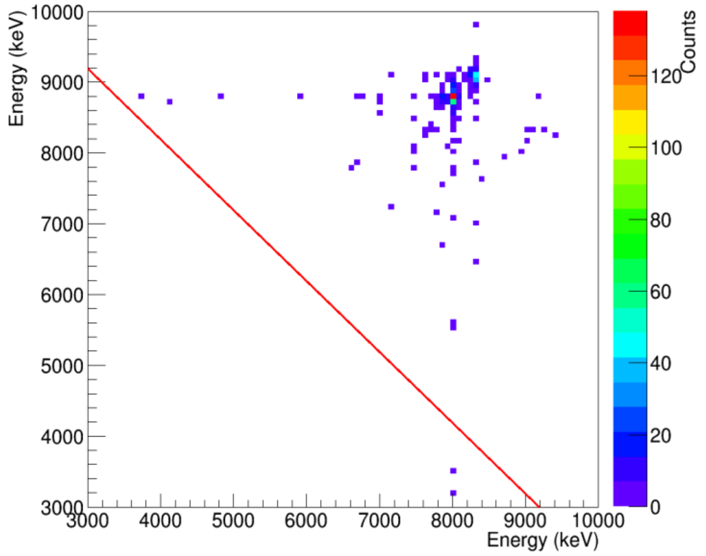
- Back to back detection, cone 4 deg. half angle (not exactly back to back + beam spot size)

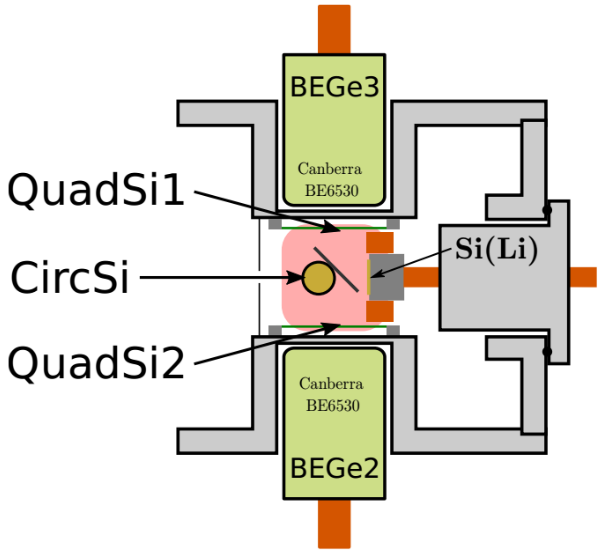
  350 Hz → 0.43 Hz

Random coincidences, selection, etc.

- Condition E_{sum} ~ 12.19 MeV

  Estimated from a ^{224}Ra experiment performed at IGISOL, JYFL





Background reduction ~ 10^4 - 10^5 (but hard to estimate more precisely).

- Pilup / pixel → negligible

True/False double-alpha detection ratio ~ 5

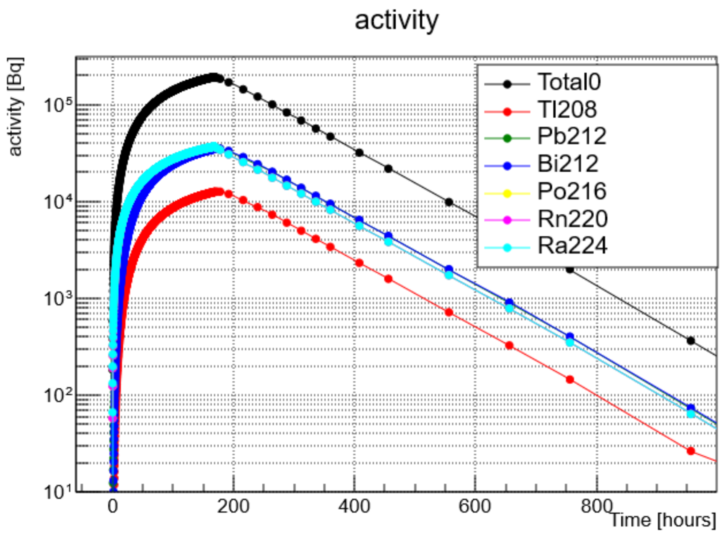
(if 10^5 E_{sum} background reduction)

Statistics (estimates)

Figures provided in the proposal should not be considered as precise numbers.

Several factors can help reducing the random correlations rate :

- Total counting rate is not constant. Background estimates are based on a maximum 3 10^5 Bq activity
- Narrow (< 100 ns) coincidence time window at the data analysis level
- Nuclei ejected from the catcher during single alpha emission
- ^{220}Rn outgassing
- Predicted branching ratio is 1.8 10^{-9}



→ random coincidence rate of 0.43 Hz is pessimistic.

→ True/False double-alpha detection ratio ~ 5 is likely a realistic estimate

TAC comments

^{224}Fr contamination

→ ^{224}Fr decays 100% toward ^{224}Ra, therefore pretty good !

^{224}RaF beam

→ We have no objections a priori; half-life modification with a molecule is unlikely

How often would foils be exchanged ?

→ never if possible

- We should avoid changing or breaking a foil (breaking would be a disaster because of the long 3.4 d ^{224}Ra half-life)
- Backup foils will be prepared in case of…
- Mechanism to insert a fresh foil will be studied, in order to monitor the implantation rate

Summary

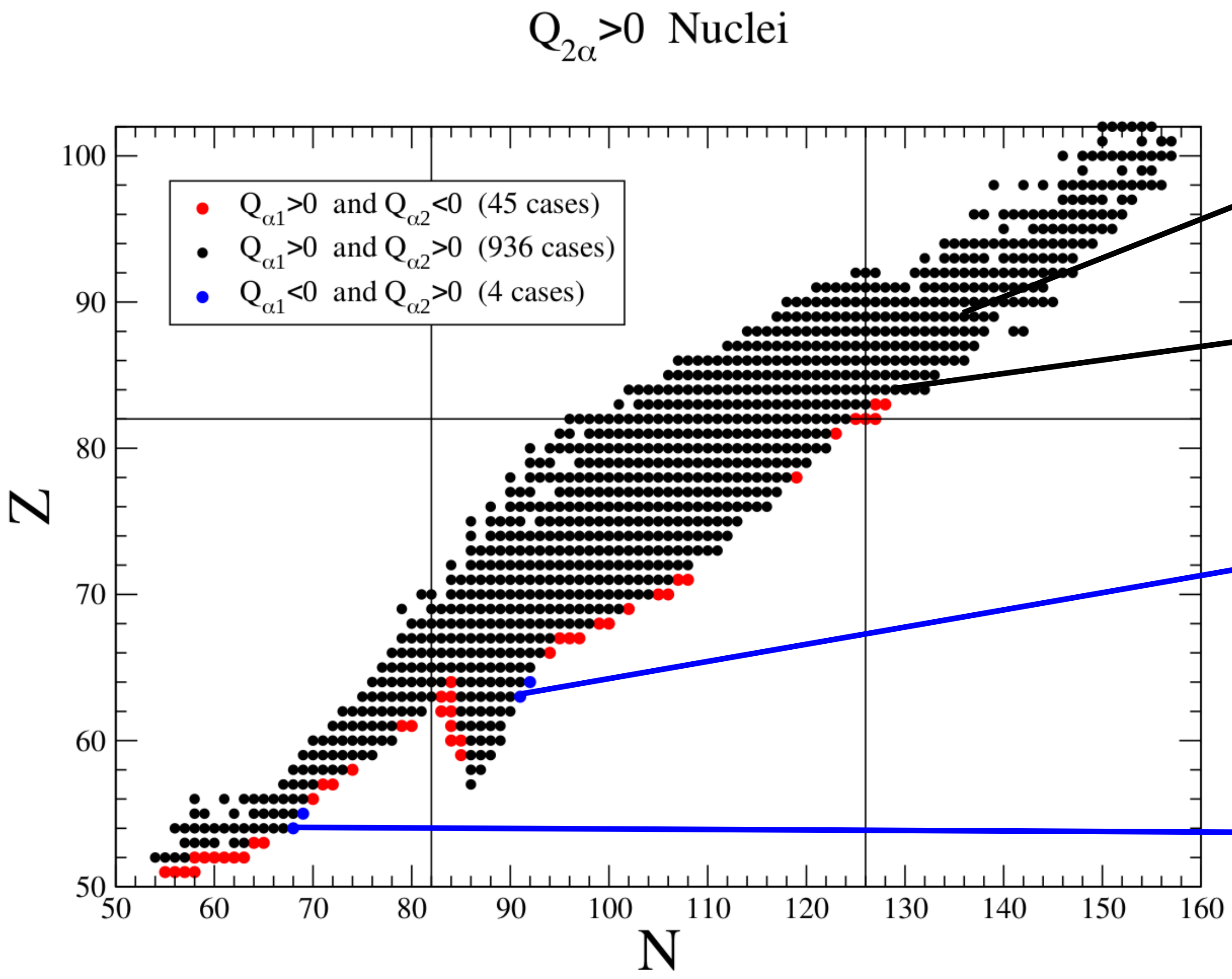
- Search for a new radioactivity mode predicted by recent theoretical calculations
- ^{224}Ra candidate selected
- $I = 5.\ 10^4$ pps, 1 week
- Si segmented detectors box MUST2/MUSETT, LA1 line
- Low statistics, huge background → challenging experiment
- Team familiar with high risk / high gain experiment
- Technical support including CEA Irfu office at CERN

Request :

- 2 shifts tuning
- 21 shifts data taking
- At least four days off-line, one week would be better

BACKUP

What about 2α emission in other nuclei ?



$\tau_{2\alpha}^{224}\mathrm{Ra} \approx 10^{14.24}\,\mathrm{s}$

$\tau_{2\alpha}^{212}\mathrm{Po} \approx 10^{18.36}\,\mathrm{s}$

$\tau_{2\alpha}^{156}\mathrm{Gd} \approx 10^{102.62}\,\mathrm{s}$

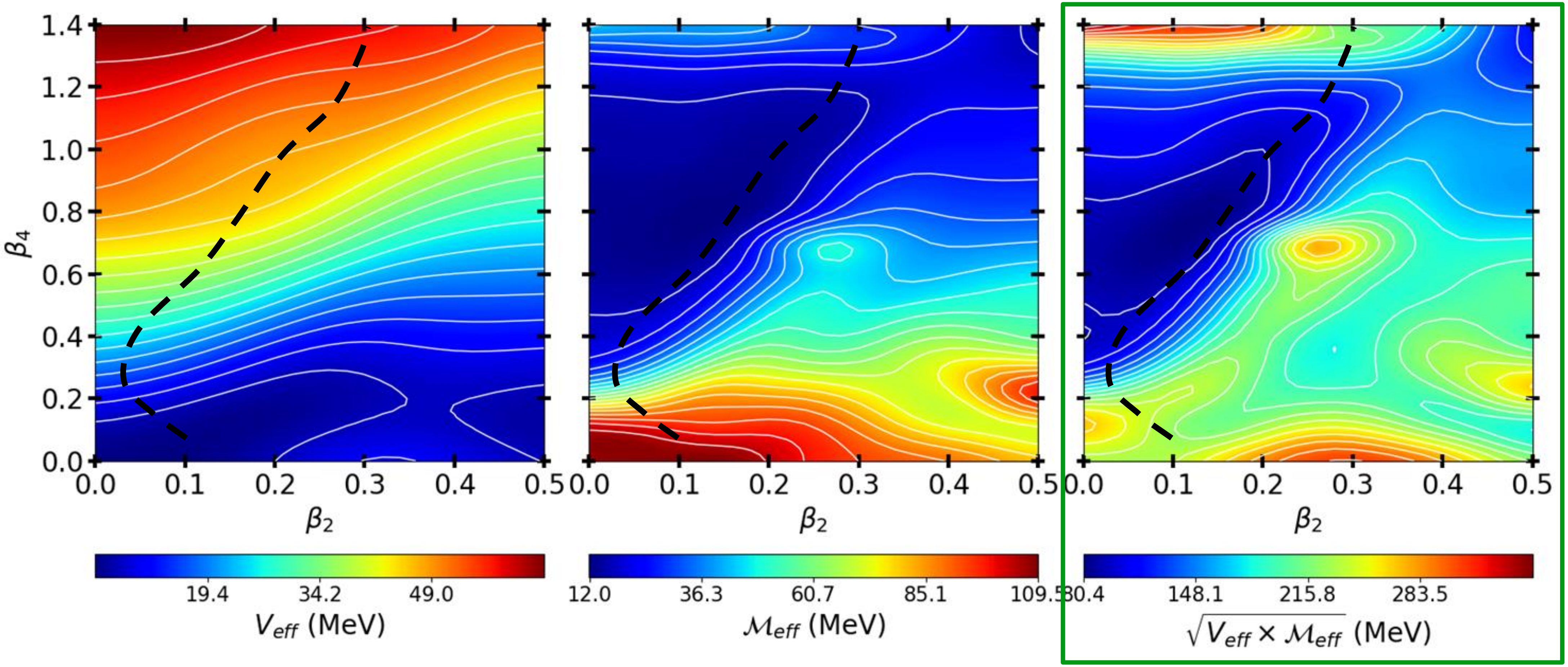
$\tau_{2\alpha}^{122}\mathrm{Xe} \approx 10^{67.46}\,\mathrm{s}$

Which constraints for each type

	Fission	Cluster	Alpha	Double alpha
β_{20}	X	X	X	X
β_{30}	X	X	X (light)	
β_{40}			X (heavy)	X

^{224}Ra : 2α emission

$$S(L) = \int_{s_{in}}^{s_{out}} \frac{1}{\hbar}\sqrt{2\mathcal{M}_{\text{eff}}(s)\left[V_{\text{eff}}(s) - E_0\right]}ds$$



Lifetime computation

What do we need to minimize to find the “good” path ?

$$S(L) = \int_{s_{\text{in}}}^{s_{\text{out}}} \frac{1}{\hbar}\sqrt{2\mathcal{M}_{\text{eff}}(s)\,[V_{\text{eff}}(s) - E_0]}\,ds$$

Inertial (effective) mass : information about the dynamic. Computed using Adiabatic Time Dependent Hartree Fock Bogoliubov method (ATDHFB) and perturbative cranking approximation

PES : information about the energy cost of a given path

$$\mathcal{M}_{\text{eff}} = \hbar^2 M_{(1)}^{-1} M_{(3)} M_{(1)}^{-1}$$

$$\left[M_{(k)}\right]_{ij} = \sum_{\mu\nu} \frac{\langle 0|\hat{Q}_i|\mu\nu\rangle\langle\mu\nu|\hat{Q}_j|0\rangle}{(E_\mu+E_\nu)^k}$$

$$\delta S = 0 \Longrightarrow \tau \approx A\exp[2S(L)]$$

WKB

$X \rightarrow Y + \alpha + \alpha'$
$Q2\alpha = mX - mY - 2m\alpha$

Conservation :
$p\alpha - p\alpha' + py = 0$
$mX = mY + 2m\alpha + E_Y + E\alpha + E\alpha'$

$E_Y = m\alpha/mY\ (E\alpha + E\alpha') - 2\ m\alpha/mY \sqrt{E\alpha E\alpha'}$
$Q2\alpha = E\alpha + E\alpha' + m\alpha/mY\ (\sqrt{E\alpha} - \sqrt{E\alpha'})^2$
$Q2\alpha = E\alpha + E\alpha'$ + (small) correction

R=Eα/Eα’	**Eα' (MeV)**	**Eα (MeV)**	**E_Y (MeV)**
0,01	11,89	0,12	0,177
0,05	11,49	0,57	0,127
0,10	11,00	1,10	0,094
0,20	10,11	2,02	0,057
0,30	9,35	2,80	0,035
0,40	8,69	3,48	0,022
0,50	8,12	4,06	0,013
0,60	7,61	4,57	0,007
0,70	7,17	5,02	0,004
0,80	6,77	5,42	0,001
0,90	6,42	5,77	0,000
1,00	6,10	6,10	0,000

^{224}Ra 30 keV in Si

^{224}Ra : Eα = 5.67 MeV → E_{Rn} = 103 keV
→ large part of Rn will leave the catcher

Range ^{224}Ra in C = 184 Å (SRIM) = 0.0184 µm
+ Rn outgassing ?

JYFL, IGISOL. p+232Th @ 65 MeV. 5 days, 7 masses settings A = 219, 221, 222, 224, 225, 226 et 228